MARS: A radioactive material beam line at Synchrotron SOLEIL

J-B. Pruvost[1]

1: Synchrotron SOLEIL – L'Orme des Merisiers, BP.48, 91192 Gif sur Yvette Cedex France.

Abstract

The Synchrotron SOLEIL is the French third generation 2.75 GeV synchrotron light source in Saint-Aubin, France. SOLEIL is now operating 25 beam lines at 430 mA and top-up mode, since December 2009.
This paper describes the MARS beam line installed on a bending magnet of SOLEIL's storage ring and the radiation safety aspects involved by the radioactive samples. Containment, shielding and dosimetry issues are presented for both the sample inside the beam line areas and external exposure in the experimental hall. This paper is mainly focusing on the containment and shielding aspects of the samples and the experimental stations.

1. Introduction

The synchrotron SOLEIL is the new French third generation synchrotron light source. It will operate 28 beam lines by the end of 2014, including 19 insertion devices and 7 bending magnet beam lines, plus 2 infra red beam lines. Twenty two beam lines (16 ID, 6 BM) plus two infra red beam lines are now operational, delivering a 430 mA synchrotron beam in top-up mode for user experimentations. One other ID beam line is under commissioning at the moment of this paper and three others are under construction.

2. Presentation of the MARS beam line

The MARS beam line is dedicated to serve a large community of scientists in the fundamental and applied research fields related to radioactive matter including Chemistry, Physics, Surface Science, Environmental Science and Life Science. This means that a very large variety of samples have to be considered as well as different techniques of characterization. The MARS beam line synchrotron light source is coming from a bending magnet of 1.71 T with a critical energy of 8.6 keV. The optics of the beam line have been optimized in order to deliver the maximum monochromatic flux inside the energy range of 3.5 to 35 keV with about 10^{12} ph/s at 12 keV and with the ability of focusing the X-ray beam to 300 x 300 μm^2 on the sample.
The MARS beam line is able to host a very large range of experiences and covers a lot of scientific topics like solution chemistry (organic and inorganic ligands and natural organic matter); solid state physics and chemistry (structural properties of nuclear fuel, matrix for nuclear waste storage, stability of actinides alloys, study of material for current and future nuclear facilities); interface chemistry (radionuclides retention in nuclear waste storage context, radionuclide migration in the geosphere) and biology (retention and migration of radionuclides in nuclear waste storage context, study of complexes of medical interest, biological effects due to radiations, nuclear toxicology studies).
These experiments are performed with different technics available on the two end station like high resolution diffraction spectroscopy (HRXRD), absorption spectroscopy (XAS, TXRD) and X-ray fluorescence (XRF) and with a very versatile sample environment capability divided in five experimental families.

- F0: refers to normal conditions with no external constraint on the sample (normal atmospheric pressure, normal temperature);
- F1: High temperature and pressure experiments up to 2000K and up to 100 GPa;
- F2: Low temperature experiments using a cryostat system bringing sample temperature in a range between 10K to 300K;
- F3: Is a family for chemical and redox reactions on the sample during synchrotron X-ray exposition mainly on liquid samples with a temperature range from 300K to 450K;

- F4: is a family dedicated to high temperature experiments from 300K to 1800K.

The total activity of the samples able to be hosted on MARS beam line at the same time is limited up to 185 GBq with a maximum activity per sample also limited to 18.5 GBq.

The beam line display in the experimental hall consist in two lead "classically" shielded hutches for synchrotron radiations with one optical hutch and one experimental hutches housing two different experimental set-up and detectors for a diffraction station and for an absorption spectroscopy end-station.
The figure 1 presents the MARS beam line display in SOLEIL's experimental hall, showing basically the three main functional areas. The first one corresponds to the 5 mm lead shielded optics hutch housing primary optics and double crystal monochromator. The second area includes the 3 mm lead shielded experimental hutch, three airlocks, a changing room and different technical rooms. The last third area consists on the main control room.
Due to the risks coming from radioactive samples to be studied and the high levels of radioactivity foreseen, the walls, the ceilings, the floors and the doors were constructed using materials and paints which can be easily cleaned (in case of contamination) and fireproof, with a level of resistance of two hours.

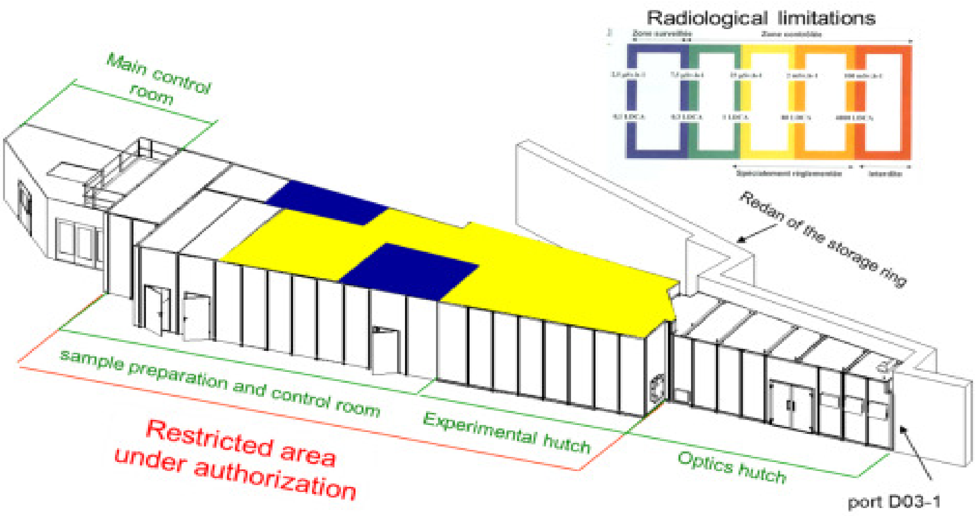


Fig. 1 – General layout of the MARS beamline from the optics hutch (right side) to the control room (left side).

The depressurized atmosphere is ensured by a dedicated ventilation system which is particularly important to dynamically contain the beam line room atmospheres in the eventuality of a contamination incident. The gradient of negative pressures from the synchrotron hall to the experimental hutch as well as the associated levels of air renewals were successfully achieved demonstrating thus the suitable air tightness of the infrastructure which is mandatory for all duties with contaminant samples. Typically, room air pressures of around -80 Pa with five complete volume renewals of air per hour have been obtained inside both preparation and experimental hutches.
Figure 2 below is showing a top-view of the MARS beam line rooms dedicated to house radioactive samples and some auxiliary technical rooms.

A fire detection system network which is interlocked with the ventilation network controller is present in all these rooms. In case of a fire detection inside or near around the beam line, the ventilator inlet is stopped. The fire shutters inserted inside the air tube are closed as well as the fire resistant doors and the fire shutter in front of the Be windows of the experimental hutch.

The main specific equipment installed that characterizes the MARS beam line infrastructure is shown on figure 3.

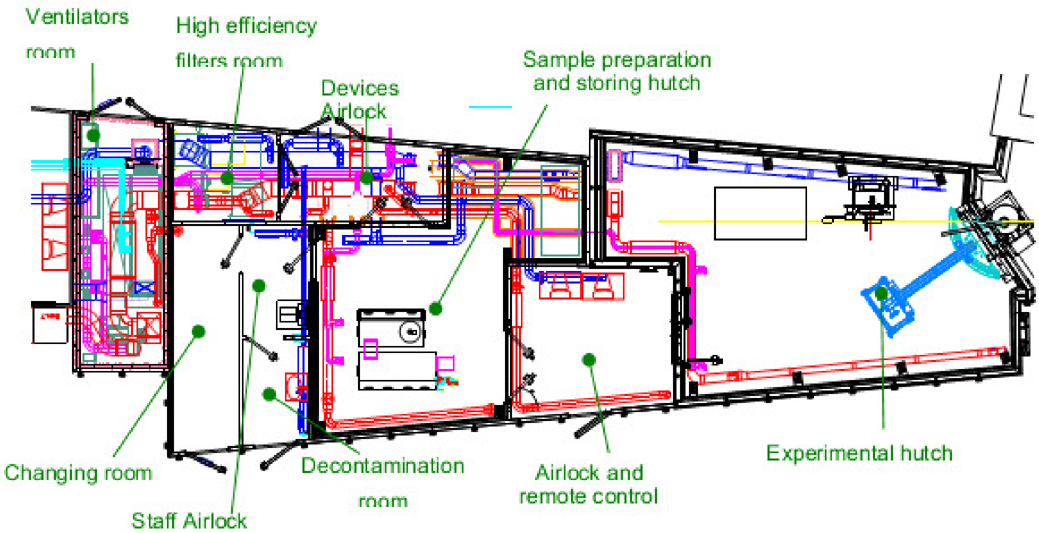


Fig. 2 – Top view of the MARS beam line showing the different hutches, the main equipment and the different pipe networks of the ventilations system.

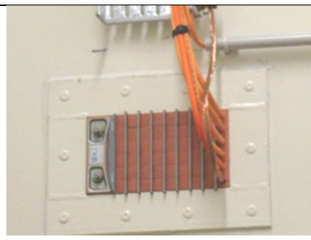
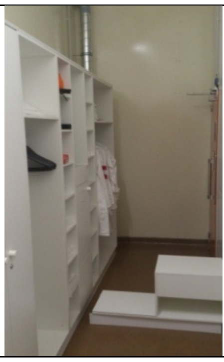
 		
Special fireproof layer on metallic walls and ceilings	Fireproof airtight feed-through for cables and pipes	Glued metal sheet, special paint and separate gantry supporting the pipes and cables networks
 		
Complex fire detection system connected with ventilation system and able to close several fire shutters and fire doors	Absolute filters on both ventilation networks and outlet ventilators connected to external chimney located on the top of the synchrotron building	
 		
Changing room and a primary decontamination safety block	Dynamic containment from two independent ventilation networks (breathable air and process)	

Fig. 3 – Specific equipment required on the MARS beam line.

The MARS beam line infrastructure is also equipped with special security system which includes a closed circuit surveillance video network (4 cameras), an access control by badges for each external doors and an emergency alarm warning.
Radiation protection devices are also present on each working area inside the rooms where radioactive samples are manipulated.
Two radioprotection controls area are present inside the sample preparation room (cf. fig. 1 & 2). One simple desk for contamination controls of the sample holders, typically for low activity samples, and one glove box for contamination controls of the sample holders including medium and high activity samples. Both working stations are equipped with emergency calling network system (pedals or button calling) and continuous air area monitoring system for radioactive aerosols detection (alpha and beta particles emitters).
Figure 4 is showing the ICAM^{TM} probe (provided by Canberra Company) near the glove box and the calling pedal below the contamination control desk. A second ICAM probe is displayed near the experimental station where the sample holders are installed for diffraction or absorption spectroscopy. Both ICAM probes are linked together and able to be displayed by the RADACS^{TM} interface on an especially dedicated computer screen in the MARS control room.

Fig. 4 – Pedal control for emergency calling system (yellow circle) and ICAM continuous air area monitor for alpha and beta aerosols emitters (red circle).

Figure 5 presents the ICAM probe display close to the absorption spectroscopy end station inside the experimental hutch.

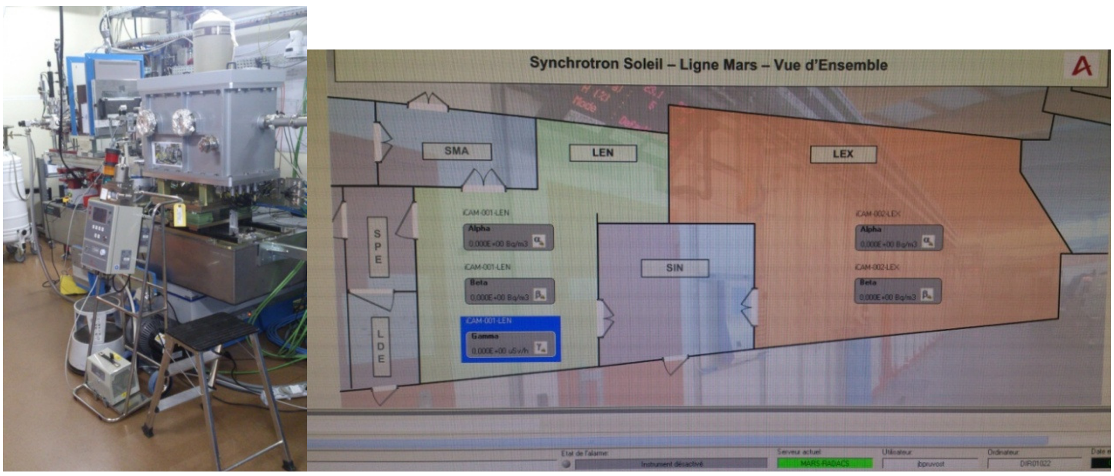


Fig. 5 – ICAM probe installed close to the absorption spectroscopy end station (left) and RADACS display of the air area control for alpha and beta emitters from the two ICAM probes (right).

The ICAM probe is mounted on a movable frame and is able to be displayed close to the experimental end station used by the scientists. On figure 5, one can see also the screen displayed by the RADACS

network system showing the alpha and beta air contamination measurements in the preparation and experimental rooms.
When radioactive samples contain gamma emitters with significant dose rate, personal dosimeters are mandatory to monitor external exposure of the staff. The personal dosimeters are EPD Mk2 type with Hp(10) and Hp(0.07) measurement capabilities both for dose rate and integrated dose of X and gamma photons and β electrons. These electronic operational personal dosimeters are provided in France by APVL Company.

Fig. 6 – Personal dosimeters with access panel installed in front of the MARS beam line control area entrance (left), personal hands and feet surface contamination LB147 monitor(Berthold) installed in the staff airlock room (center), hands and cloths contamination monitor (Canberra's MIP10+SAB100 probe) (right).

The radiation safety group attributes to each person involved in an experiment with radioactive samples a specific individual dosimeter with a personal code number. Individual exposures are recorded in a database and send periodically to the Radiation Safety French Authority.
Figure 6 presents the different radiation protection materials deployed for personal exposure and contamination controls.
For personal control, each working station is also equipped with a portable alpha/beta hands and cloths controller. Before exiting controlled area, it is mandatory for any staff member or scientific users to control themselves with hands and feet surface contamination monitor installed in the exiting staff airlock room prior to access the changing room.

3. The radioprotection policy at SOLEIL

The general radiation safety goal at SOLEIL is that all SOLEIL staff and users are considered as non-classified workers regarding ionizing radiation hazards in the experimental hall. This means that in all normal operation conditions, the radiation doses outside the accelerator tunnels or outside the beam line hutches are below the maximum dose limit authorized for non-classified people. That means annual individual effective dose below 1 mSv. This is translated as an operational goal for the design of the shielding as an average dose rate limit of 0.5 $\mu Sv.h^{-1}$.
The welcome on MARS beam line of experiments on radioactive samples should have no effect on SOLEIL operations for both accelerators and beam lines operation at any time. So, whatever the radioactivity level welcomed on MARS, equivalent dose rates outside the beam line rooms have to be below 0.5 $\mu Sv.h^{-1}$.
Direct contact between SOLEL staff or users and the radioactive samples is strictly forbidden and all sample preparation has to be done in user's laboratory prior to come in SOLEIL.
For the MARS operation durability no contamination hazard is affordable, so a very particular attention as to be dedicated on sample containment by users. Containment has to be designed and tested by users to ensure efficiency during transport and experiment at SOLEIL in all conditions. Results are controlled and approved by SOLEIL radiation safety group before experiment acceptance. Then official clearance

is only given for the experiment at SOLEIL after the acceptation of the project by the French Regulation Authority.
Annual individual dose objective is to stay below 2 mSv per year for MARS beam line staff and radiation safety group members as well as for the other SOLEIL staff members.

4 Radioactive samples hosted on MARS beam line

A very large amount of different samples of any kind is able to be studied at MARS beam line, representing about 400 different isotopes with activity limited up to 18.5 GBq per sample or sample holder. Only solid and liquid samples are allowed on MARS beam line, including sintered powders but easily spreadable samples like gaseous or powder samples are strictly forbidden.
The main limitation in order to accept samples on MARS beam line is the contamination levels in case of a leakage of the sample containment and the possible impact in case of dispersion for the workers, the facility and environment. Limitation on sample is also coming from the dose rate levels they are able to generate outside the beam line limits in the experimental hall.
In order to give an idea of the diversity of sample type to be analyzed on MARS beam line here is some typical examples below:

- Doped glass for nuclear waste management studies, and long live and high activity waste studies (with sample typical radioactivity from 100 kBq to 2 GBq);
- Environment samples, solid in solution and liquid, for geological or biological studies (mainly Th and/or U isotopes from few Bq to about 2 kBq per sample);
- Structure material in nuclear power plants (activated metallic alloys samples) with typical radioactivity levels from few kBq to 10 GBq;
- New and spent fuel samples like sections of UO_2 fuel pellets up to 3 GBq and 23 mg.

5 Containment and shielding

If contamination risk is present because of the sample itself and or because of the experience, then three independent containment barriers are mandatory between the sample and the environment.
Two independent barriers are required directly around the sample and generally part of the sample holder. A third independent barrier is present between the sample and the environment thanks to the beam line airtight rooms and the air depression cascade between inside and outside.
Several types of containment have been developed by users in order to give a safe confinement of the radioactive samples and to allow the primary synchrotron beam to scan the sample and the secondary light (transmitted, diffracted radiation or fluorescent emission) to go through the sample holder and reach dedicated detectors with a minimum of signal absorption.
As a first example, PEEK[1] dome sample holders are used for diffraction experiments and mainly for solid samples of low activity level and for normal conditions experiments (F0 family). Up to four different samples can be loaded inside, with for each of them a kapton[2] tape as first containment barrier.
Figure 7 shows a PEEK dome sample holder (from Brucker) for diffraction experiments and how it can be loaded with four different samples sealed in kapton tape.

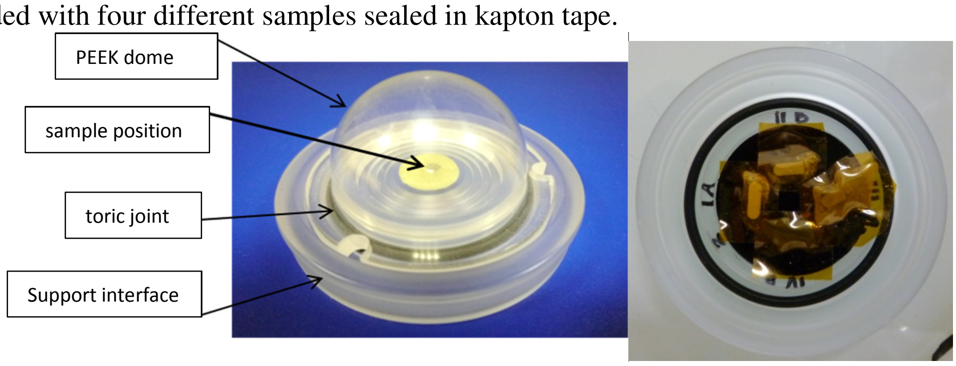


Fig. 7 – PEEK dome sample holder with mounting of four different samples, each one sealed in a kapton tape.

[1] polyethertherketone
[2] Polyimide film with silicon adhesive

For more specific environment experiments, special sample holders have been developed in collaboration with user's laboratories like, for example, the sample holder designed for electrochemical experiments (F3 family) as shown on figure 8 below. The external barrier is designed with 3 kapton windows for the incoming synchrotron X-rays, the transmitted X-rays and the fluorescence emission X-rays respectively.

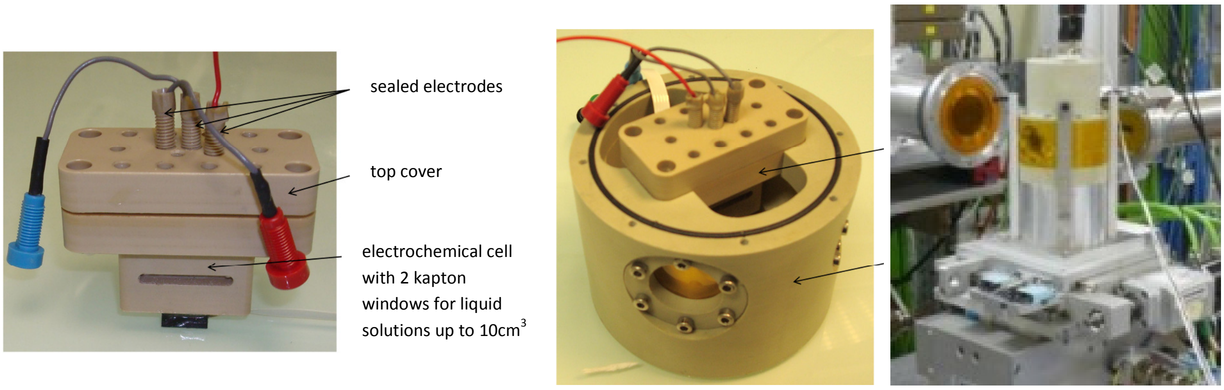


Fig. 8 – Special sample holder for F3 family (oxide-reduction) experiments; the first barrier electrochemical cell (on left), the cell mounted on the bottom part of the second barrier (center) and the complete setup installed on the XAS-XRF end station of MARS beam line.

In order to limit the number of sample replacements per experimental project, multi samples holders have been developed for both solid and liquid samples with a loading capability up to twelve different samples. As an example, the figure 9 presents a multi samples holder for F0 family experiments able to be loaded with 12 samples of different activities and or concentrations (if liquids) and generally used for low activity samples (range: 10 to 10^3 Bq/sample) like natural or depleted uranium samples or thorium samples from environmental studies.

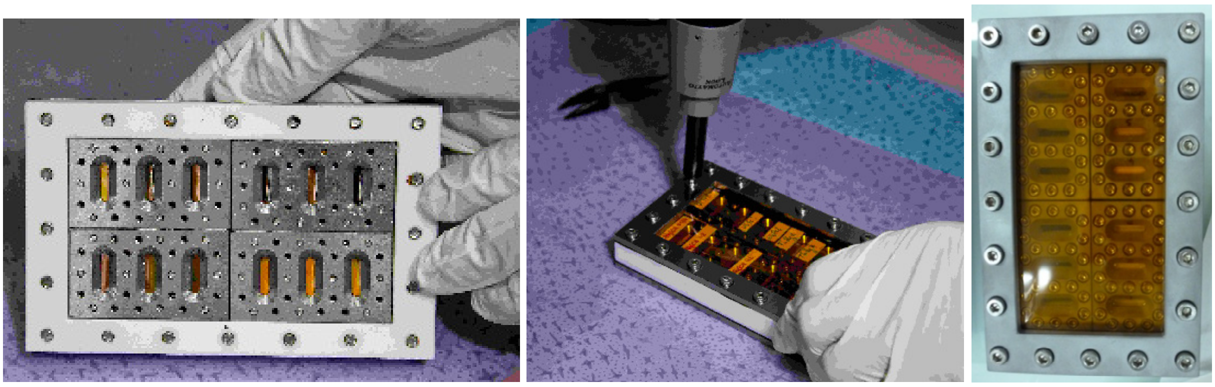


Fig. 9 – Mounting of a sample holder for F0 family experiments with solid or liquid samples with 12 individual cells for sample.

It is composed of four different Teflon[3] elements with three cells sealed by a Teflon cap with Viton joint allowing the filling of the cell and two 200 µm thick Teflon windows both sides mounted on a Teflon block with a stainless steel frame, each element composing the first barrier. Then these four elements are sealed together in another Teflon block sealed with once again on both side a 200 µm Teflon sheet with a flat Viton joint and a stainless steel frame screwed all around.

For high activity samples (range of few MBq up to 18.5 GBq), another type of specific containment has been developed for F0 and F4 families of experiments. It is mainly dedicated to nuclear fuel pellet samples coming from PWR UO_2 fuel element but is also able to host, for example, activated metallic alloys samples coming from several structural materials of different types of nuclear power plants.

[3] polytetrafluoroethylene

Due to the high dose rate levels generated by these samples, activity of spent fuel pellets able to be studied on MARS beam line is limited to about 3 GBq which will induce anyway gamma dose rate levels of about 200 $\mu Sv.h^{-1}$ at 1 m (Hp(10) and about 2 $mSv.h^{-1}$ at 30 cm) and beta dose rate levels of about 100 $mSv.h^{-1}$ at 30 cm (Hp(0.07)).
This is for this reason that PEEK dome was substituted by Beryllium dome for the sample holder, particularly because of the high beta dose rate effect on the PEEK dome, damaging the material stability. Figure 10 presents the sample holder for this kind of high activity sample, based on a Russian puppet design as shown on the picture below.

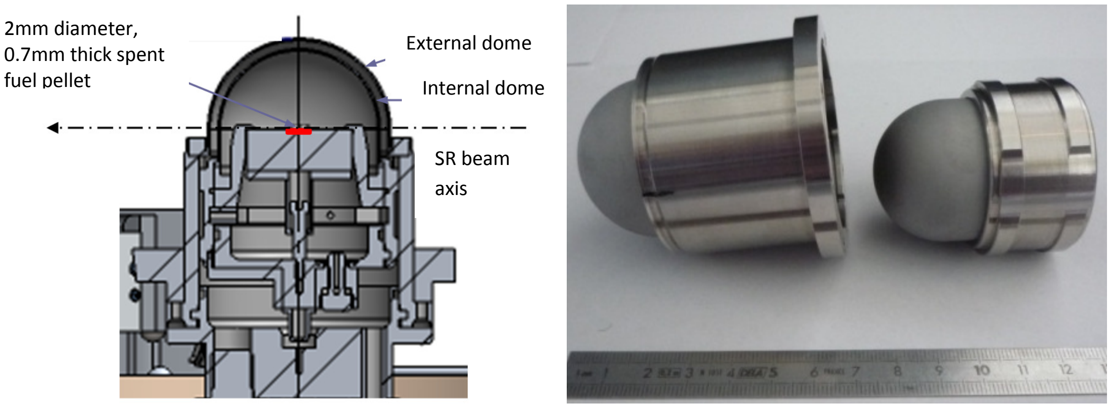


Fig. 10 – Sample holder for nuclear fuel pellet, brand new or spent fuel up to 90 GW.d/t after at least one to five years of cooling and decay.

With such sample, direct contact with the sample holder is not allowed at SOLEIL, so for transfer and radiological controls, the sample holder is manipulated with a special and long three digit tweezers. Thus, it is necessary to have a shielding around the sample holder during the whole experiment, from its arrival at MARS beam line storage and preparation room and to its whole stay on the analyzing end station. For this reason, a carrying shielding is under design for the transfer operations from the storage room to the experimental end station and back. Figure 11 below presents the transfer tweezers and a scheme of the carrying shielding made of at least 3 cm thick lead walls.

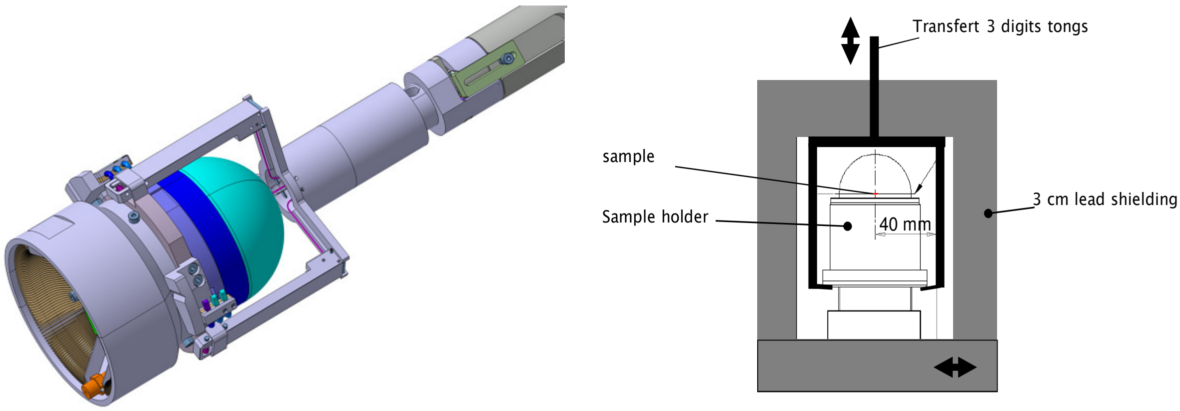


Fig. 11 – New and spent fuel sample holder tweezers and transfer shielding.

Because of the high dose rates generated by the sample it is necessary to protect both personal staff and detector from the radiation. For this reason, the sample is never directly visible from the detector but always behind a shielding. The transmitted emission from the sample due to the synchrotron light is deflected by an analyzer crystal towards the detector as shown on the scheme on the left side of figure 12 below.

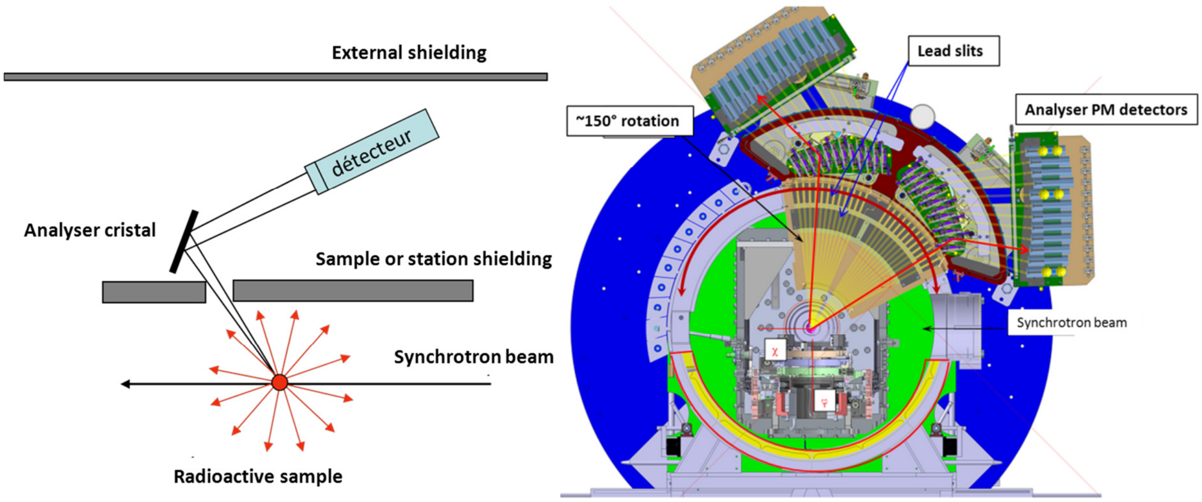


Fig. 12 – Basic scheme (on the left) of the measurement technic to avoid high background on the detector coming from the radioactive sample radiations and development of this principle for the shielding of the diffraction end station.

But it is also mandatory, to meet the dose rate threshold levels outside the limits of the MARS beam line rooms, to have an additional shielding around the two experimental end stations. Figure 13 presents the end station shielding for both the diffraction and absorption experimental stations.

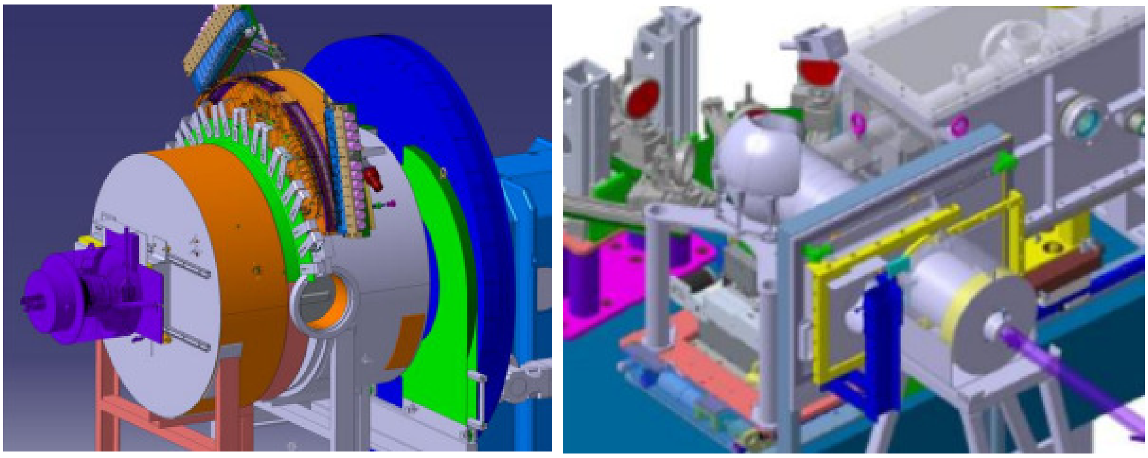


Fig. 13 –Left picture: shielding of the diffraction end station showing the integration of the multianalyzer detector (on the top) and the transfer shielding docked on it (on the front in purple). Right picture: shielding of the absorption end station with the integration of the multi crystal spectrometer (on the back) and the transfer shielding docked in front with tweezers extension stick (on the right in purple).

In addition with these end station shieldings, it was also mandatory to add another additional shielding alongside of the experimental hutch wall because, when the shielding aperture is open to allow X-rays on the detector, the necessary shielding for the highest dose rate emitter samples is far away thicker than the requested shielding of the hutch toward synchrotron scattered X-rays (about 6 cm of lead instead of 3 mm respectively) in order to respect the ambiance dose rate threshold of the experimental hall (0.5 $\mu Sv.h^{-1}$).
A lead wall with variable thickness (from 2 cm to 6.5 cm thick) as a function of the horizontal angle and distance from the two end stations was set alongside of the experimental hutch side wall as shown on the picture 14 below.

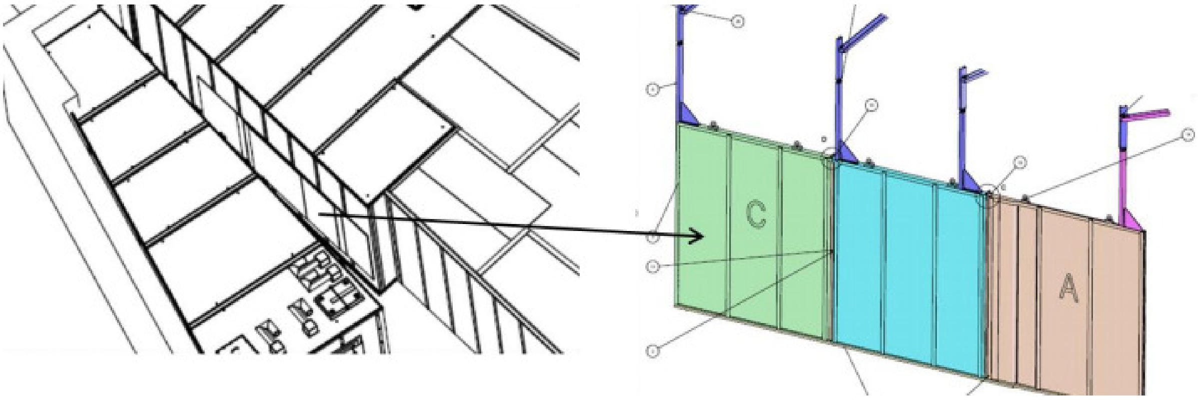


Fig. 14 – Representation of the additional shielding that was installed on the side wall of MARS experimental hutch.

For the storage of the samples during their stay at SOLEIL a safe storage has been foreseen in the preparation and storage room of the MARS beam line. Assuming a maximum of six spent fuel samples at the same time, each with their individual transfer shielding of 3 cm of lead, a shielded safe storage has been designed. Main calculations were done with MERCURAD™ code (from AREVA) and lead of the needs for 5 cm of lead wall panels and doors and a 6.5 cm of lead for the side wall panel as shown on figure 15 below.

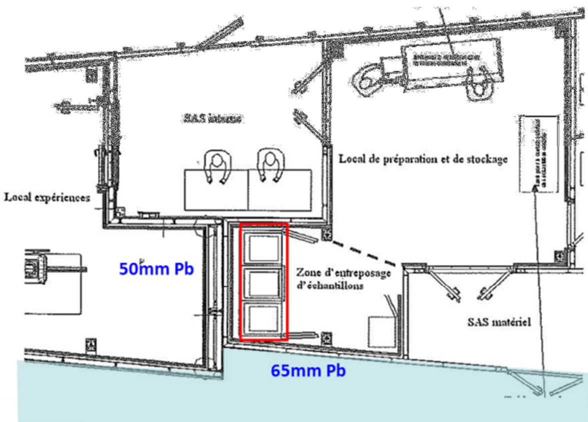


Fig. 15 –The safe storage (in red) on the MARS beam line and required lead thickness of its shielding.

5 Status and perspectives

The MARS beam line is already running operation with radioactive samples below French exemption limit thresholds since 2010 with a specific authorization for a limited number of experiments on only 20 different isotopes (instead of 400) from the French Regulation Authority (ASN). Table 1 below presents the different experimental families currently authorized and the different authorization steps expected from the ASN for the authorization of the operation of the MARS beam line with radioactive samples.

Radioactive classes of samples		Technological fields: F0	F1	F2	F3	F4
	C0 (A <1 ET)	green	green	orange	green	green
	C1 (A <200 ET)	yellow	orange	orange	orange	orange
	C2 (A <2 10^4 ET)	yellow	grey	red	red	red
	C3 (A <2 10^6 ET)	red	grey	grey	grey	red

- Green: Currently authorized
- Yellow: 1^{er} level of authorization
- Orange: 2^{ème} level of authorization
- Red: 3^{ème} level of authorization

Nevertheless, a little bit more of 37 experiments with radioactive samples were achieved at the moment of this paper was written, welcoming about 250 users and, for the experiments finished in 2012, with satisfying scientific results and publications. Nominal operation with radioactive samples is foreseen before the end of 2015 representing about 100 samples analysed and 28 experiments per year.